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Growth of Ge hexagonal meshwork films on Si(111)-7 × 7

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Abstract
Ge growth on a Si(111)-7 × 7 surface at room temperature was studied by *in situ* scanning tunnelling microscopy. The Ge hexagonal meshwork film composed of Ge nanoclusters located on the centre of the half-unit-cells of Si(111)-7 × 7 grows in the 'monolayer mode': the first layer is 0.22 nm thick, and then increases in thickness by 0.11 nm, forming the 0.33 nm bilayer. This growth mode leads to a simple stacking sequence. However, the Stranski–Krastanov growth mode was still found for the thick meshwork films.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Growth of Ge on Si(111) has attracted much interest both for fundamental research and technical applications in photo-electronics and low-dimensional devices. Usually, the Stranski–Krastanov (SK) growth mode occurs for Ge growth on Si(111) due to the big lattice mismatch between Ge and Si (4.2%) [1, 2]: the Ge first grows in the layer-by-layer mode for a few monolayers (ML), and then three-dimensional (3D) islands develop on the preforming wetting layer. Both (7 × 7) and (5 × 5) reconstructions were observed for thin Ge films grown on Si(111)-7 × 7 at elevated temperatures [2–4]. Recently, a kind of hexagonal meshwork superstructure of Ge on the Si(111)-7 × 7 surface grown at room temperature (RT) was observed [5, 6]. Each hexagon is composed of six Ge nanoclusters located on the centres of six adjacent half-unit-cells (HUCs) surrounding the corner hole of the Si(111)-7 × 7 surface. Very recently, Ansari *et al* also studied the formation process of small Ge clusters on Si(111)-7 × 7 [7]. However, to our knowledge, no other studies on the hexagonal meshwork evolution have been done since it was first found. So, some basic questions remain about the hexagonal meshwork structure. For instance, how does the hexagonal meshwork develop if more Ge is deposited? Since such a structure is obtained at RT, is there any correlation between it and the monolayer growth mode found at RT in Ge growth on Ge(111) or Si on Si(111) early [8–10], and if so, what?

In this paper, we report on our studies on the hexagonal meshwork evolution as a function of Ge coverage on the Si(111)-7 × 7 surface at RT. The so-called 'monolayer growth mode' was found for the 2D Ge meshwork films. The detailed growth process of the monolayer mode and of the stacking sequence was proposed accordingly. In this case, the Ge film still follows the SK growth mode.

2. Experimental details

The experiments were performed in an ultrahigh vacuum (UHV) system equipped with a variable temperature scanning tunnel microscope (VT-STM) (Omicron, Germany). The base pressure is better than 1×10^{-10} mbar. The clean Si(111)-7 × 7 surface was obtained in the UHV system by first degassing at about 600 °C for more than 12 h and then quickly flashing to 1250 °C by direct current (DC) heating followed by slowly cooling down to room temperature at a rate of ∼2 °C s^{-1}. High purity Ge (99.9999%) was sublimed onto the clean Si(111) surface by DC heating with a typical deposition rate of ∼0.01 ML min^{-1}. Here, 1 ML is defined as the atomic density of the Si(111)-1 × 1 structure. During the deposition, the Si(111) substrates were kept at RT. Chemically etched tungsten tips were used for scanning. The empty- and filled-state STM images were acquired in the constant current mode at RT.

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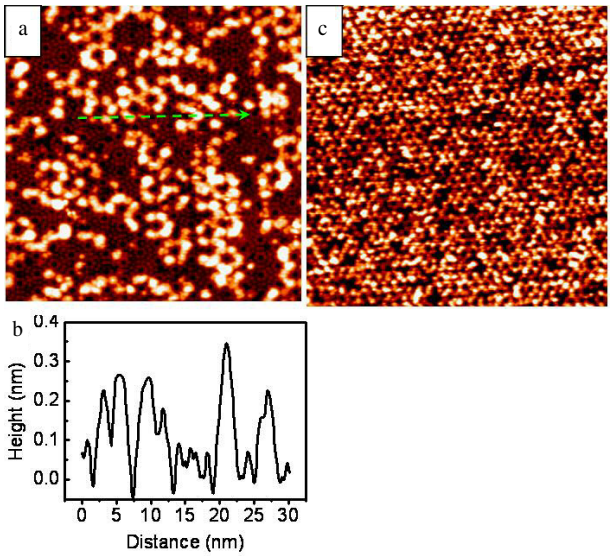

Figure 1. (a) Empty-state STM image (3.21 V, 0.054 nA) of about 0.2 ML Ge coverage on Si(111)-7 × 7. The area is 50 × 50 nm^2; (b) the height line profile across the clusters marked by the dashed arrow line shown in (a); (c) empty-state STM image (2.60 V, 0.022 nA) of about 0.4 ML Ge coverage on Si(111)-7 × 7. The area is 100 × 100 nm^2.

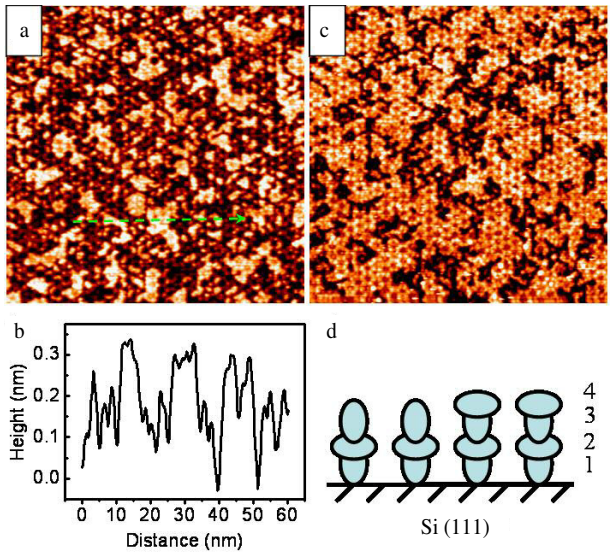

Figure 2. (a) Empty-state STM image (3.21 V, 0.034 nA) of about 0.5 ML Ge coverage on Si(111)-7 × 7. The area is 100 × 100 nm^2; (b) the height line profile across the islands marked by the dashed arrow line shown in (a); (c) empty-state STM image (2.60 V, 0.037 nA) of about 2.0 ML Ge coverage on Si(111)-7 × 7. The area is 100 × 100 nm^2; (d) side view of the schematic drawing of the growth process of Ge hexagonal meshwork films on Si(111).

3. Results and discussion

Figure 1(a) shows an empty-state STM image of about 0.2 ML Ge coverage on the Si(111)-7 × 7 surface. The Ge clusters with different sizes and shapes are distributed irregularly, and the separate clusters and those forming dendritic structures coexist. This indicates that at RT the irregular Ge clusters are located randomly on the Si(111) surface. But careful observation reveals that all the randomly distributed Ge clusters actually occupy centres of the HUCs of the Si(111)-7 × 7 reconstructed surface. This means that all the Ge clusters are still confined in the HUC despite their different sizes. It is noted that, except for the very small cluster looking like an individual Ge atom, no atomic resolution was obtained for the normal Ge clusters on the image. This is probably because the bondings between Ge atoms within each cluster or between the Ge atom and the Si atom on the substrate surface are not so strong that the Ge atoms have higher activities.

Figure 1(b) shows the height line profile across the clusters shown in figure 1(a) marked by the dashed arrow line. On average, the height of most Ge clusters is about 0.22 nm, and that of the big cluster is 0.33 nm. Since each cluster occupies one HUC, then, if all the HUCs on the Si(111)-7 × 7 surface are occupied by Ge clusters, a kind of hexagonal meshwork structure will form. In fact, this structure has already been observed by others previously [5, 6]. Figure 1(c) shows an STM image of about 0.4 ML Ge coverage on Si(111)-7 × 7 at RT. The hexagonal meshwork structure was clearly observed. Such a structure remains stable when the annealing temperature is up to 100 °C. The clusters forming the hexagonal structure are not identical, but most are about 0.22 nm high. This is similar to that of 0.2 ML Ge coverage.

It is well known that at moderate temperature, Ge film grows epitaxially on Si(111) with a diamond-shaped structure. And then the question arises of how does the meshwork structure develop at RT if more Ge is deposited.

To answer the question, the empty-state STM image of 0.5 ML Ge coverage is presented in figure 2(a). Besides the finished first Ge layer of about 0.22 nm thickness, some 2D islands composed of a similar hexagonal structure are also observed. The height profile through the dashed lines in figure 2(a) is shown in figure 2(b). The clusters in the islands are higher than the clusters in the finished first layer by about 0.11 nm. This value is the same as the height difference between the smaller and the bigger Ge clusters shown in figure 1(a), meaning that the 0.11 nm high islands (actually including the nether parts) are composed of 0.33 nm high Ge clusters.

Accordingly, an interesting growth process of the Ge meshwork films is proposed as follows: first, the 0.22 nm thick layer forms, and then it increases in thickness by 0.11 nm so that a 0.33 nm thick bilayer forms. That is to say, in the cluster scale, the 0.22 nm high clusters first form and occupy all the HUCs of the Si(111)-7 × 7 surface, and then the Ge clusters start increasing their sizes with the additional height of 0.11 nm. When all the clusters become big (0.33 nm in height), the 0.33 nm thick Ge cluster bilayer finishes growing. In fact, we also noted that the surface morphology of the 0.22 nm thick Ge cluster layer is a little different from that of the 0.33 nm thick Ge cluster layer. After the 0.33 nm thick Ge cluster bilayer is complete, new 0.22 nm high Ge clusters nucleate and occupy the top of the big clusters in the finished bilayer, meaning that the second bilayer is developing. The side view of the schematic drawing of the growth process is shown in figure 2(d). Here, '1' denotes the first monolayer formed by the small clusters adsorbed on Si(111); '1' and '2' together denote the first bilayer formed by the big clusters. Similarly,

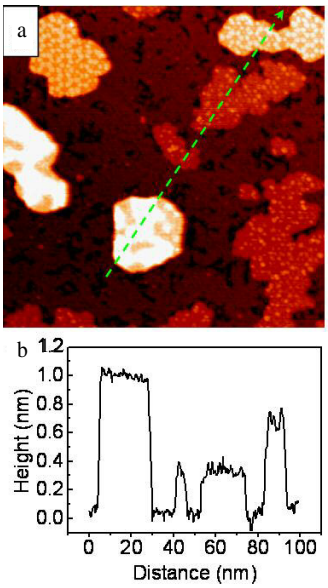

Figure 3. (a) Filled-state STM image (−3.21 V, 0.031 nA) of about 4.0 ML Ge coverage on Si(111)-7 × 7. The area is 100 × 100 nm^2; (b) the height line profile across the islands marked by the dashed arrow line shown in (a).

'3' and '4' respectively represent the third and the fourth monolayer, and together represent the second bilayer. The growth process stated as above should be the essential of the so-called monolayer growth mode found also in Ge/Ge(111) and Si/Si(111) at RT mainly by reflection high energy electron diffraction [8–10]. Such a growth mode with a simplest stacking configuration leads to a honeycomb structure built with the hollow centred hexagonal meshwork for the thick Ge cluster film. Obviously, the atomic density of this meshwork film is much less than that of the diamond structure.

The STM image of ∼2.0 ML coverage of Ge is presented in figure 2(c). The hexagonal meshwork is clearly displayed on the top layer. This means that the Ge meshwork film still grows in the 2D layer-by-layer growth mode on Si(111) at RT for the Ge coverage at least up to 2.0 ML.

The filled-state STM image for ∼4.0 ML Ge coverage is presented in figure 3(a). Obviously, in this case, besides the 2D wetting layer and 2D islands, some 3D islands with different heights appear. This demonstrates that Ge cluster film grows on Si(111)-7 × 7 at RT, still taking advantage of the SK growth mode. This also indicates that the strain caused by the lattice mismatch between Ge and Si diamond crystal structure still exists within the Ge hexagonal meshwork layers although the hexagonal meshwork has less atomic density than the usual diamond structure.

The height profile of the 2D and 3D islands is shown in figure 3(b). It is expected that the heights of the islands are about an integer times 3.3 Å. The analysis of the refined structure on the protuberance of the line profile shows that the same monolayer growth mode occurs for the islands as for the films. This accounts for the fact that two kinds of hexagonal meshwork structures are both observed on the 2D wetting layer or islands and on the 3D islands.

Although without atomic resolution for the clusters, from the values of the cluster sizes we can conceive some information of their structure configurations. For instance, 0.33 nm is approximately equal to the distance (3.2 Å) between the adjacent (111) planes of Ge diamond structured single crystal. And 0.22 nm is close to the bond length of Si–Si or Ge–Ge in the diamond structure (2.3–2.4 Å). Considering that the distance (3.2 Å) between the adjacent (111) planes of Ge diamond structure consists of one vertical Ge–Ge bond (2.4 Å) and one slantwise Ge–Ge bond (0.8 Å), we propose that for the small cluster in the first layer, Ge atoms form nearly vertical bonds with the Si atoms within the HUCs, probably the Si adatoms; for the big clusters, the upper Ge atoms bond with the underlying Ge atoms inside the small clusters, inclining about 60° from the Si(111) surface.

4. Conclusion

In conclusion, Ge growth on the Si(111)-7 × 7 surface at RT was studied by *in situ* VT-STM. The Ge hexagonal meshwork superstructure film grows in the monolayer mode: the first layer is 0.22 nm thick, and then it increases in thickness by 0.11 nm, forming one bilayer. For the 0.22 nm thick layer, the clusters located on the centre of the HUCs of Si(111)-7 × 7 are 0.22 nm in height; and for the 0.33 nm thick bilayer, 0.33 nm. That is to say, the 0.22 nm high clusters first form and occupy all the HUCs of the Si(111)-7 × 7 surface, and then the clusters start increasing their sizes with the additional height of 0.11 nm. When all the clusters become big (0.33 nm in height), the 0.33 nm thick bilayer finishes growing. After that, the new small clusters start to nucleate on the centre of the underlying big clusters again, meaning the second bilayer develops. This growth mode leads to a simple stacking sequence. In addition, the SK growth mode still occurs for the thick meshwork films.

Acknowledgments

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